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Abstract

There is an urgent need to improve the reliability of screening cargo containers for illicit nuclear material that may be hidden there for terrorist purposes. A screening system is described for detection of fissionable material hidden in maritime cargo containers. The system makes use of a low intensity neutron beam for producing fission; and the detection of the abundant high-energy γ rays emitted in the β-decay of short-lived fission products and β-delayed neutrons. The abundance of the delayed γ rays is almost an order of magnitude larger than that of the delayed neutrons normally used to detect fission and they are emitted on about the same time scale as the delayed neutrons, i.e., ~1 min. The energy and temporal distributions of the delayed γ rays provide a unique signature of fission.

Because of their high energy, these delayed γ rays penetrate low-Z cargoes much more readily than the delayed neutrons. Coupled with their higher abundance, the signal from the delayed γ rays escaping from the container is predicted to be as much as six decades more intense than the delayed neutron signal, depending upon the type and thickness of the intervening cargo.

The γ rays are detected in a large array of scintillators located along the sides of the container as it is moved through them. Measurements have confirmed the signal strength in somewhat idealized experiments and have also identified one interference when 14.5 MeV neutrons from the D, T reaction are used for the interrogation. The interference can be removed easily by the appropriate choice of the neutron source.

Introduction

During the year 2001 more than six million maritime cargo containers were delivered to US ports[1]. Half of them came from the top ten foreign ports of origin and nearly 90% arrived at the top ten US ports[1]. The capacity of these containers can exceed 25,000 kg of cargo, which could provide a convenient hiding place for illicit delivery of weapons of mass destruction or their components to the US. Current plans call for detection of special nuclear materials (SNM) that might be hidden in these containers with passive radiation detectors sensitive to the normal neutrons and γ rays emitted in the radioactive decay of these components. However, the normal radiations emitted from plutonium can be attenuated readily by cargo materials and therefore the passive detection of plutonium may not be as reliable as desired. Similar considerations apply to the attenuation of the normal radiations emitted by ^{235}U. In this case the only radiation of significant intensity is a γ ray at 186 keV that is readily attenuated in a loaded cargo container

The radiation signatures of uranium and plutonium can be modified and increased in intensity by interrogating these targets with neutrons or γ rays to produce fission. Following fission, both prompt neutrons and γ rays are released. At much later times as a result of β–decay of the fission products, very characteristic delayed neutrons[2-5] and γ rays are emitted. The prompt radiations are hard to detect because of the overwhelming abundance of neutrons and γ rays produced by the interrogation beam. While delayed neutrons can be easily distinguished from beam neutrons, they have relatively low yield in fission, approximately 0.008 per fission in ^{239}Pu and 0.017 per fission in ^{235}U[6]. Further, they are rapidly attenuated in hydrogenous materials. Consequently, traditional passive and active techniques for detection of SNM are unreliable at best in the presence of thick cargo of hydrogenous materials that might conceal the illicit SNM.

Utilizing the high-energy γ-ray signature of fission products

There are many fission products with relatively high yield that have short half-lives and thus high specific activities. Some of them produce γ-rays with energies exceeding 2.5 MeV. These high-energy delayed γ-rays have nearly ten times greater yield than the delayed neutrons[7], as has been confirmed by recent experiments[8]. Their high-energy makes this γ-radiation a characteristic of fission, very distinct from normal radioactive background that typically produces no γ-radiation exceeding an energy of 2.6 MeV. The present work utilizes detection of this distinctive γ-ray signature following neutron-induced fission to detect SNM hidden in cargo containers. The result is the so-called “nuclear car wash” illustrated schematically in Figure 1 below.

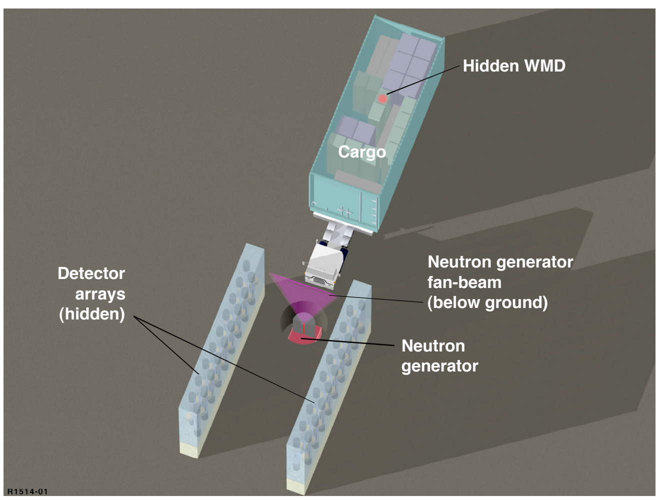

Figure 1 Schematic of the “nuclear car wash” [7] showing a below-ground collimated neutron source irradiating the container from below, and two linear detector arrays to detect subsequent fission product β-delayed γ-rays.

In the figure a neutron generator is buried below ground and its output collimated into a thin fan at the ground surface. That fan beam illuminates the cargo and produces fission in SNM that may be hidden there. The use of a neutron beam for interrogation is not necessary. A high-energy photon source ($E_\gamma > 6$ MeV) could also be used with similar effect. The cargo container is towed at constant velocity over the neutron source then through a cell with two linear arrays of γ-ray and neutron detectors on the sides, analogous to a car wash. The detector array is configured to distinguish between photons with energies above and below 2.5 MeV. Decay of fission products will be observed as a bright spot moving along the detector array at the speed of the container. The fission products have a mixture of decay times but the principal intensity decays with a half-life of approximately 25-55 s [7, 8], depending on the irradiation pulse length, and this characteristic can be used to distinguish SNM signature radiation from neutron activation products.

In the approach described here, the high-energy γ-ray signature for SNM is superior to that of delayed neutrons in the case where the intervening cargo material is hydrogenous due to its much higher penetrability as compared to low energy delayed neutrons. This is seen in Figure 2.

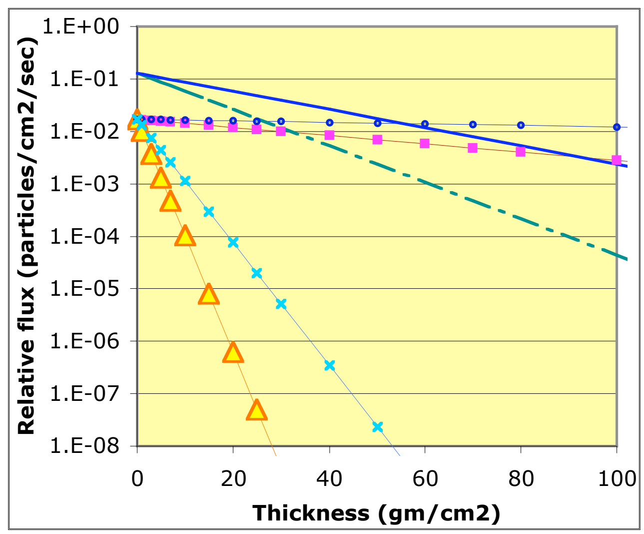

Figure 2. Attenuation of neutrons and high-energy γ-rays in various media. Solid line is 3 MeV γ-rays in water. Dot-dash line is 3 MeV γ-rays in Pb. Solid circles are 300 keV neutrons in Pb, Boxes are 300 keV neutrons in Al. X is 300 keV neutrons in wood. Triangles are 300 keV neutrons in water.

The total yield of β-delayed neutrons and high-energy ($E_\gamma > 3$ MeV) γ-rays has been estimated[7] and fixes the left hand intercept in the figure. Then Monte Carlo simulations were used to determine the effective attenuation coefficients for neutrons and γ-rays in aluminum, water, wood, and lead. The attenuated signal strength is plotted in the figure as a function of intervening cargo thickness.

Examination of the figure shows that the high-energy γ-ray intensity escaping the cargo exceeds the delayed neutron intensity for all materials to a thickness of ~ 30 gm/cm^2. In the case of hydrogenous materials such as wood and water the γ-radiation escaping the cargo greatly exceeds the neutron intensity. The actual benefit in using this signature depends on the type of material and its thickness, but can range up to six orders of magnitude. At ~ 60 gm/cm^2, a typical half-thickness for a fully loaded cargo container, the attenuated γ-radiation exceeds the neutron intensity by more than four orders of magnitude. Conversely, if the intervening material is a metal such as aluminum or lead the neutron penetration is good and exceeds the γ-ray intensity for thickness exceeding 40-80 gm/cm^2 of metal. Thus the concept proposed above includes detection of both neutrons and high-energy γ-radiation.

Confirmation of γ-ray spectra and identification of interferences

A collimated 14 MeV neutron generator with 2×10^{10} n/s output produced a neutron beam that irradiated a standard 20 ft cargo container from outside its sidewall. A moderated target of 22 kg natural uranium (150 g ^{235}U) was placed inside on its centerline. The target uranium was in the form of ~ 3 mm diameter pellets, filling a polyethylene jar of dimensions 10 cm diameter and 18 cm length. This target construction allowed several different containers to be used, providing useful variations in the geometric shape of the target and its thickness. It was located in the beam at a distance 2.2 m from the neutron source, and 1.3 m from the wall of the cargo container. The target was immersed in polyethylene beads forming a moderator of thickness 13 cm that completely surrounded the target. Simulations confirmed that nearly all of the fissions were caused by thermal neutrons in this configuration.

Photon emission from the target was monitored using a well-collimated and shielded HPGe γ-ray spectrometer placed inside the cargo container. Its efficiency was 50% relative to 3x3 NaI detector and it was located immediately outside and approximately normal to the collimated neutron beam at a distance 1 m from the target.

The neutron generator was pulsed with a 50 % duty factor with cycle times ranging from 60 s (30 s on and 30 s off) to 200 s. Spectral measurements of γ-rays were made between neutron pulses in “event mode”. The γ-ray pulse-height data were sorted off-line to produce spectra of events recorded during particular time ranges following the end of the neutron pulse. Generally, data were accumulated over a period of ~3-5 hours of interrogation, i.e. ~300 beam pulse cycles.

Examples of the γ-ray pulse-height spectra are shown in Figure 3 below.

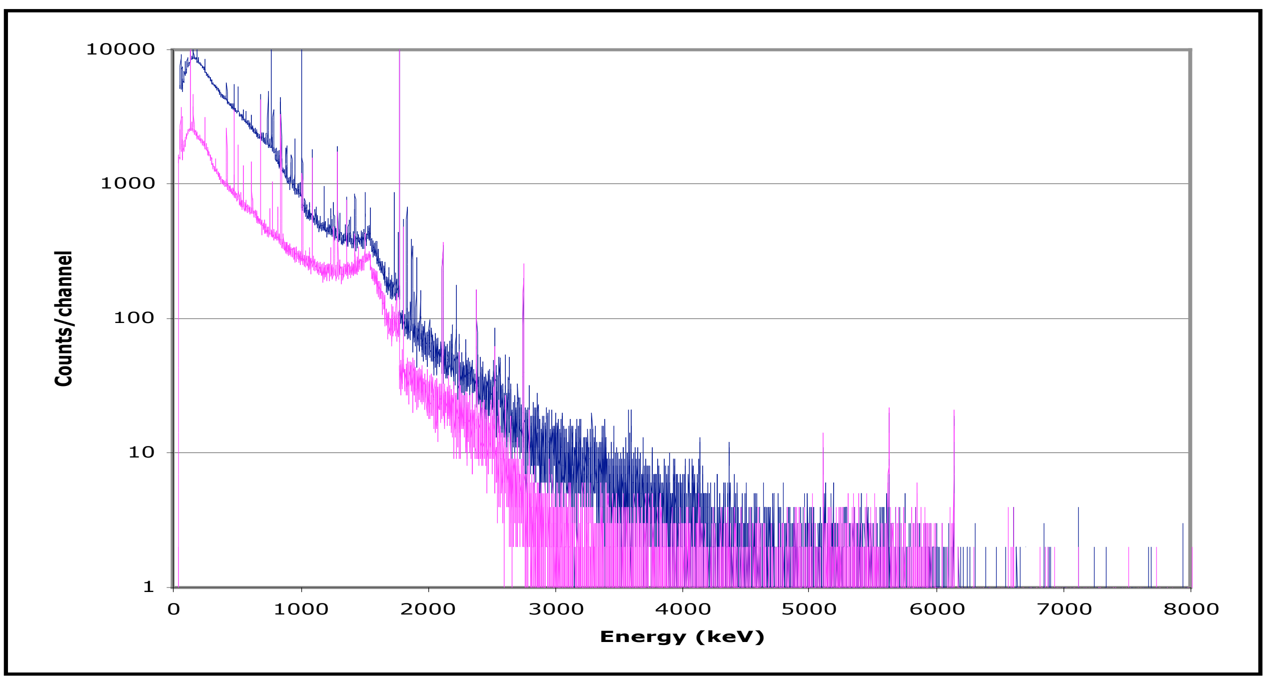

Figure 3. Pulse height spectrum with and without a natural uranium target. Data shown in blue (upper) were accumulated between neutron beam pulses with the target in place. Data shown in red (lower, background) were accumulated between neutron beam pulses with the same experimental arrangement except that the target replaced by an empty polyethylene container.

The data shown were taken with a 200 s pulsing period. The data shown in blue (upper) were taken with the uranium target present, and the data shown in red (lower) were taken with exactly the same experimental arrangement except for the absence of the target. Examination of the figure shows that there is substantial γ-ray intensity due to the target at all energies. Many of the intense lines observed have been assigned to high-yield fission products such as $^{86-88}$Br, $^{90-92}$Rb [9]. The most prominent spectral feature that is due to the presence of the uranium target is the relatively high intensity in the energy range 2-4 MeV where the target produces more than five times greater intensity than is present in the neutron activated background. The high-energy part of the spectrum can be seen more clearly in the expanded view shown in Figure 4 below.

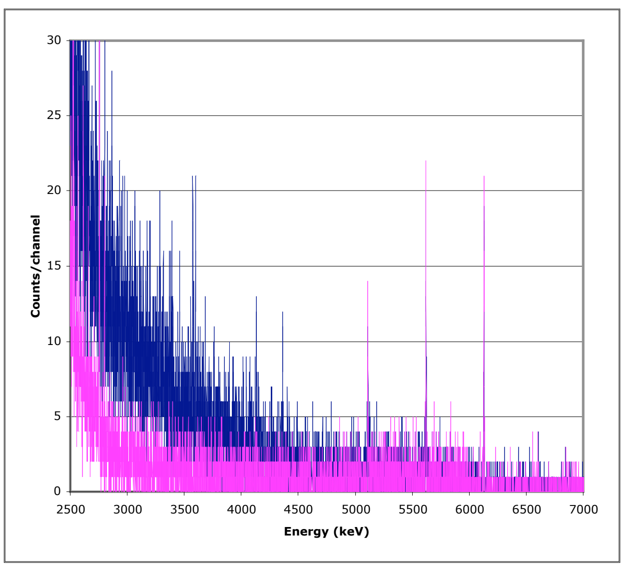

Figure 4. Expanded view of the data in Figure 3 showing high-energy fission product γ-rays and ^{16}N decay.

In addition to the target γ-rays, both the foreground and background pulse height spectra exhibit strong radiation at 6.1 MeV, along with its single- and double-escape peaks, and Compton distribution. This is due to the decay of 7.16-s ^{16}N. Interrogation with 14.5 MeV neutrons results in the reaction ^{16}O(n,p)^{16}N, which has an effective threshold of E_n=10 MeV.

The γ-ray pulse-height data were collected as sequential spectra each of 1 s duration beginning at the end of the neutron pulse to permit a study of the temporal behavior of the high-energy delayed γ-rays. In Figure 5 are shown decay curves constructed by integrating the total intensity in each of the 1-s spectra over the energy range $E_\gamma \geq 2.5$ MeV.

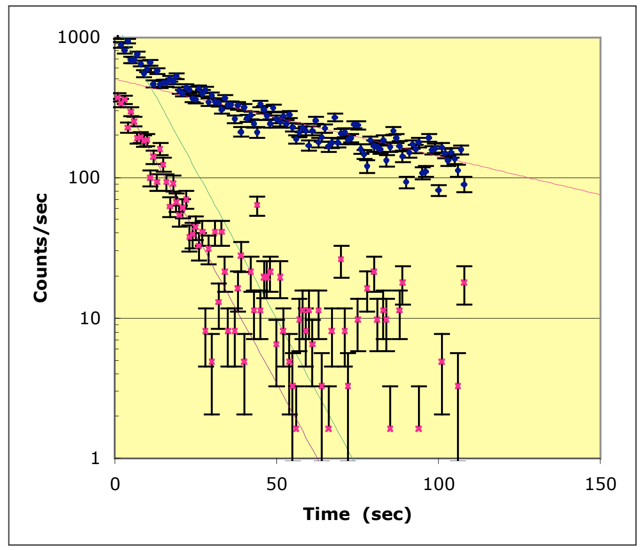

Figure 5. Decay of γ-radiation at energies above 2.5 MeV for a 200-s interrogation cycle. Two data sets are shown: solid circles are foreground, i.e. target present, and X are background, i.e. target removed. Lines are not fits but intended to guide the eye. They correspond to 7.1 and 55 s half-lives.

Examination of the figure shows that the high-energy portion of the background is dominated by the decay of ^{16}N for approximately 1 min following the end of the beam pulse. There is no compelling evidence for the presence of any other activated species that contributes to the high-energy background. The high-energy portion of the spectrum in the presence of the uranium target is also dominated by the decay of ^{16}N for the first 15 s following the end of the beam pulse, but the remaining time interval is dominated by decay with a gross average half-life of about 55 s. This half-life can be understood in terms of the fission products whose high-energy γ-ray yield are highest in ^{235}U fission, i.e. 55-s ^{86}Br, 55 s-^{87}Br, 156-s ^{90}Rb, 258-s ^{90m}Rb and 58-s ^{91}Rb[7]. Further, some of the intensity attributed to ^{16}N must be due to the decay of a number of shorter-lived fission products such as 4.35-s ^{89}Br, 4.49-s ^{92}Rb, etc. Because of the wide range of the half-lives of the fission products, it is expected that the observed decay times will depend on the length of the beam pulse and the length of the subsequent observation period.

Conclusion

A concept for detection of SNM hidden in cargo containers is proposed and some of its elements studied. The method is based on neutron interrogation to produce fission followed by detection of β-delayed high-energy γ radiation and/or delayed neutrons from decay of the fission products. The concept includes irradiation of a cargo container by a collimated neutron source located below ground. The entire contents of the container are irradiated as the container passes over the source. The container then passes through a linear array of neutron and γ-ray detectors where both delayed neutrons and delayed γ rays are detected.

Because the β-delayed high-energy γ rays are nearly a decade more abundant than delayed neutrons, tend to have much higher penetrabilities through common low-Z materials, and because the energy spectrum and decay properties of the high-energy delayed γ rays are a characteristic signature of fission, it is expected that this signal will dominate the detected events in most cases. Estimates of attenuation in normal cargos indicate that the high-energy γ-ray signature of fission products escaping from thick hydrogenous cargo may be up to six decades larger than the β–delayed neutron signal, depending on the type and thickness of intervening material

When 14.5 MeV neutrons are used as the interrogating source, interference from oxygen activation can be expected. This can be eliminated by interrogation at a neutron energy less than about 10 MeV. With that improvement the signal to background ratio is expected to be a great deal larger. In the presence of the oxygen interference the fission product signal is still expected to be dominant at times longer than about 15 s after the end of the neutron beam pulse

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